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PB-299 358

Analytical Diffusion Model for Long Distance Transport of Air Pollutants

Massachusetts Inst of Tech, Cambridge Fluid Mechanics Lab

Prepared for

Environmental Sciences Research Lab, Research Triangle Park, NC

Jun 79

United States
Environmental Protection
Agency

Environmental Sciences Research
Laboratory
Research Triangle Park NC 27711

EPA-600/4-79-037
June 1979

Research and Development

PB 299358

EPA

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TECHNICAL REPORT DATA
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1. REPORT NO. EPA-600/4-79-037	2.
3. RECIPIENT'S ACCESSION NO. PB299358	
4. TITLE AND SUBTITLE ANALYTICAL DIFFUSION MODEL FOR LONG DISTANCE TRANSPORT OF AIR POLLUTANTS	5. REPORT DATE June 1979 6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) James A. Fay and Jacob J. Rosenzweig	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS Fluid Mechanics Laboratory Department of Mechanical Engineering Massachusetts Institute of Technology Cambridge, MA 02139	10. PROGRAM ELEMENT NO. 1AA603 AB-015 (FY-79) 11. CONTRACT/GRANT NO. Grant No. 804891
12. SPONSORING AGENCY NAME AND ADDRESS Environmental Sciences Research Laboratory--RTP,NC Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, NC 27711	13. TYPE OF REPORT AND PERIOD COVERED Final 14. SPONSORING AGENCY CODE EPA/600/09
15. SUPPLEMENTARY NOTES	

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17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
* Air pollution * Mathematical models * Atmospheric diffusion Deposition		13B 12A 04A

18. DISTRIBUTION STATEMENT RELEASE TO PUBLIC	19. SECURITY CLASS *(This Report)* UNCLASSIFIED 20. SECURITY CLASS *(This page)* UNCLASSIFIED	21. NO. OF PAGES 29 22. PRICE A03 - A01

EPA Form 2220-1 (9-73)

EPA-600/4-79-037
June 1979

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AFFILIATION

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ABSTRACT

A steady-state two-dimensional diffusion model suitable for predicting ambient air pollutant concentrations averaged over a long time period (e.g., month, season or year) and resulting from the transport of pollutants for distances greater than about 100 km from the source is described. Analytical solutions are derived for the primary pollutant emitted from a point source and for secondary pollutant formed from it. Depletion effects, whether due to wet or dry deposition or chemical conversion to another species, are accounted for in these models as first order processes. Thus, solutions for multiple point sources may be superimposed.

The analytical theory for the dispersion of a primary pollutant is compared with the numerical predictions of a plume trajectory model for the case of steady emission from a point source. Good overall agreement between the two models is achieved whether or not depletion by wet and dry deposition is included.

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The horizontal length scale characterizing the sulphate concentration distribution from a single source is about 500 km, being noticeably larger than that for primary (sulfur dioxide) distribution. Using optimum dispersion parameters, a point source of 33 kg/s of sulfur dioxide would give rise to a maximum annual average sulfate concentration of 1 $\mu g/m^3$.

A calculation of annual average SO_2 concentrations in the United States is carried out using previously derived optimal values of the parameters from the sulfate calculation. The resulting isopleths are similar to measured values in the eastern U.S.

SECTION 1

INTRODUCTION

The transport of air pollutants to great distances from a source becomes a matter of concern when the aggregate effects of many sources may produce harmful ambient pollutant levels or depositions far downwind (10^2 to 10^3 km) of the source region. Most attention has focussed on sulfur compounds and acidic rainfall (see Symposium on sulfur in the atmosphere, 1978), but the long distance transport of ozone (Wolff et al., 1977), photochemical oxidants and their precursors (Cleveland et al., 1976), pesticides (Glotfelty, 1978) and nitrates (National Research Council, 1978) also may have significant environmental effects far downstream of their sources. A principal difficulty in understanding quantitatively such phenomena is the limited ability of models to describe accurately the transport, dispersion, transformation and deposition at greater distances and far longer travel times than has customarily been treated in the past when examining pollutant transport close to the source. This paper describes a relatively simple analytical model which may help to provide estimates of long term average pollutant levels at large distances from a large collection of sources. It also permits a rapid assessment of the contribution to the pollutant level at a given receptor of a single source or class of sources.

In one way or another, all long distance transport models provide for the principal physical effects considered to be significant in the process: advective motion of the atmosphere, dispersion caused by atmospheric turbulence, transformation of primary to secondary pollutants, and wet and dry deposition of pollutants. In the case of sulfur compounds, all of these processes are thought to be significant, at least in some region of the flow field, and the corresponding models exhibit considerable complexity. But the understanding of the contributory processes is limited and simplifications (e.g., modelling of the transformation as a first order process) are often used.

Current long distance transport models can be classified conveniently into two categories. In the first are models for which the diffusion equation is integrated over a limited domain of time and space, such as several days and distances of the order of 10^3 km. These models are probably most useful in studying short duration episodes of high pollutant levels (Hidy et al., 1978). The second category uses a calculation of air parcel trajectories to determine advective motion, with various methods for treating the dispersion about the mean motion of the parcel. These latter models are most often used to determine ambient levels or deposition rates averaged over a period of a month to a year, and over travel distances up to 10^4 km (Eliassen, 1978, Johnson et al., 1978, Ottar, 1978).

A model of the first variety, described by Hidy et al. (1976), considers concentration variation in three dimensions and time. The governing three dimensional unsteady diffusion equation is averaged over the period of 24 hours. Depletion of pollutants is viewed as first order in pollutant concentration. Since the governing equations are solved numerically the horizontal wind specified at each grid point can approximate realistically the actual short term averaged wind field. In another model, Gillani and Husar (1976) assume a steady state diffusion equation for transport scales from 1 - 100 km. The mean wind is assumed uniform while the horizontal cross wind eddy diffusivity varies with downwind distance. Again the rate of primary pollutant loss is assumed to be first order.

Trajectory diffusion models can be further categorized as puff or plume models. Eliassen and Saltbones (1975) developed a receptor-oriented puff model. Daily trajectories terminating at a receptor point are determined backward in time for 48 hours. A parcel of air moving along the trajectory is assumed to be subjected to an instantaneous infusion of pollutant from adjacent area sources while the rate of loss of pollutant occurs as a continuous first order process. The concentration of pollutant within the air parcel is assumed uniform. Four puffs arriving at the sampling point are appropriately averaged to determine mean daily pollutant concentration. Start and Wendell (1974) developed a source-oriented trajectory puff model, where source emissions are approximated by a series of puff releases. The center of mass of each puff is determined by trajectory ends while the concentration distribution, about the center of each puff, is assumed to be Gaussian in the vertical and horizontal directions. Rates of diffusion about the puff centers may vary along the trajectory length. Concentrations at a given point are determined by superposing concentrations due to all puffs. In a similar model, Sheih (1977) assumed the concentration distribution of each puff as Gaussian in the horizontal direction. However, the vertical concentration distribution is determined by numerical integration of nonsteady diffusion equations for primary and secondary pollutants. In this case emissions from elevated sources and depletion of pollutant to the ground can be treated more appropriately.

A trajectory plume model developed by Heffter et al. (1975) was applied using length scales between one hundred and several thousand kilometers and averaging times of one month or greater. Trajectories initiated from the source four times daily are assumed to be the instantaneous centerlines for a quasi-steady Gaussian plume. Contributions from each plume are superposed and averaged over an appropriate time interval. Depletion due to various mechanisms is incorporated in the model by adjusting the source strength term appropriately. Draxler (1976) added a finite difference scheme for vertical diffusion to this trajectory plume model, thus giving a more realistic concentration profile in the vertical direction and treating wet or dry deposition as a boundary condition.

Validation of these models is limited by the sparsity of suitable field observations. The most extensive comparison of a model with field data has been that for sulfur compounds in western Europe (Ottar, 1978). Since the models take into account the major mechanisms involved in the long distance transport of pollutants, namely, the large scale variability of the wind velocity and depletion effects, they require detailed knowledge of the wind field and precipitation over large regions for the duration of the time-averaging

interval. Thus their major disadvantage is the need for substantial computer capacity in order to account for the details of meteorology.

The diffusion model discussed below differs from those mentioned above in that it does not require detailed knowledge of the history of the wind field. By averaging over a time period of one month or greater, it is assumed that the governing diffusion equations are time independent and that the wind field can be usefully approximated as being uniform over a large spatial scale. To account for the advective motions developed in detail in other (unsteady) models we employ a horizontal diffusion coefficient suitable for the large time and length scales involved.

Based upon the very limited comparison with a trajectory model, which we describe below, our model successfully describes the advective effects of the variable wind field for an averaging time of a month. When applied to a determination of annual average ambient sulfate levels in the U.S., this model (incorporating first order transformation and deposition effects) correlates the observations about as successfully as do the more elaborate trajectory models for sulfur compounds in the western European region (Ottar 1978). Because of its simplicity, this model may be a useful adjunct to the repertoire of models needed to understand better the problem of long distance transport.

In what follows, we present in Section 2 the theory and analytical solutions of our diffusion model. In Section 3 we carry out calculations for a point source and compare the results with trajectory-plume model predictions. Section 4 describes a calculation of sulfate dispersion in the United States due to emissions from electric power plants of SO_2. This calculation is compared with data and parameters are adjusted for the best possible agreement with data. Using the best values of the parameters an SO_2 calculation for the United States is also carried out.

SECTION 2

THEORY AND ANALYTICAL SOLUTIONS

As an alternative to the models described above, we propose a steady state two dimensional diffusion model to describe the long distance transport of primary and secondary air pollutants. It is assumed that primary emissions occur from an ensemble of continuous point sources. The source of secondary pollutant is distributed continuously throughout the entire region due to the volumetric conversion of primary pollutant. Both types of pollutant are assumed to mix uniformly in the vertical direction within a well-mixed region of the atmospheric boundary layer. Thus ambient concentrations vary as a function of horizontal coordinates only. Removal of pollutant occurs due to wet and dry deposition and chemical transformation to other species.

The large scale horizontal dispersal of atmospheric pollutants may be thought of as analogous to a two dimensional random walk process. A continuous source of pollutant may be viewed as a serial emitter of infinitesimal puffs, each puff being transported in a random manner by the turbulent wind fluctuations. Given that the scale of our problem is on the order of several thousand kilometers it becomes apparent that the large turbulent eddies (e.g. on the order of several hundred km.) are dominant in dispersing the puffs about the point of emission while smaller turbulent eddies are responsible for dispersing each puff about its center of mass. We neglect the latter small scale distortion of the puff and instead consider each puff as if it were an infinitesimal point mass. This simplification is appropriate in determining the spatial concentration distribution averaged over long time periods since it superposes the contributions from each puff. Thus we are interested in determining the horizontal puff dispersion primarily due to the large scale turbulent wind motions. In the limiting case of infinitely many puffs averaged over a long time our problem reduces to a solution of the steady state diffusion equation with an appropriately determined horizontal diffusion coefficient.

Taylor (1921) showed formally how a diffusion coefficient may be determined by extending the statistical analysis of the random walk process to the continuous motions of turbulent flow. We choose to avoid the formalism of Taylor's analysis but take note of the conclusion reached that for large travel times an ensemble of diffusing particles has an rms displacement about their common center of mass which is proportional to the square root of the travel time. Therefore, for large travel times the diffusion coefficient approaches a constant value, as it should for a random walk process. As a consequence, we assume a constant horizontal diffusivity D_h that is determined to an order of magnitude by the relation

$$D_h \approx V^2 \tau_p \qquad (1)$$

where V is the Eulerian standard deviation of the wind velocity and τ_p is the Eulerian mean persistence time of the wind with respect to direction, which is analogous to the mean free time between collisions of a gas molecule. An order of magnitude equivalence is assumed between the diffusivity as used in an Eulerian sense with an ensemble average of Lagrangian measurements. Although a more precise relationship for the horizontal diffusivity might be derived with great difficulty we do not now consider such an improvement. Instead, we intend to adjust τ_p or D_h in order to achieve the best agreement with other calculations or with measured data.

Limited data exists on typical values of τ_p. Shirvaikar (1972) measured at ground level the persistence time for a given direction within a sector of 22.5°. He found at several sites that the distribution of persistence time is lognormal with a standard deviation of 2 to 3 hours. Pasquill (1974) noted that the Lagrangian time scale in the crosswind direction is on the order of 5 to 15 hours. Thus we expect τ_p to be on the order of several hours. Assuming V is 5 m/s, D_h would then be about 5×10^5 m^2/s.

Estimates of D_h have been made by several investigators using geostrophic trajectory calculations. Murgatroyd (1969) used two independent methods for calculating the horizontal diffusivity. In the first method, the standard deviation σ of the end points of the trajectories launched from a fixed point were determined as a function of travel time and the corresponding diffusion coefficient, also a function of time, was determined from

$$D_h = \sigma^2/2t \qquad (2)$$

The asymptotic value of D_h was extrapolated as $t \to \infty$. The second method assumed an autocorrelation coefficient of the damped harmonic form and values of the parameters where chosen by regression with trajectory data. The correlation coefficient was then substituted into the equations derived by Taylor (1921) to determine the asymptotic value for the diffusion coefficient. The resulting calculations showed D_h to be about 2×10^6 m^2/s at the lowest elevation (700 mb). Durst et al. (1959) also used trajectories to calculate the diffusion coefficient by three independent methods and found values of D_h in the range of 10^6 to 10^7 m^2/s at 700 mb. Thus, the order of magnitude of $D_h \sim 10^6$ m^2/s seems to be well established.

Because our model describes the dispersion process averaged over a long period of time (e.g., month, season or year), time-averaged values are used for the process parameters such as precipitation rate, dry deposition rate, transformation rate, wind speed/direction, etc. A more substantial question is whether these time-averaged parameters vary also with position.

As a first approximation we have assumed that these parameters are constant over the geographic region of interest, permitting an analytical form for the solution of the horizontal diffusion equation. An alternative, which we have not attempted, is to consider these parameters to be position dependent and obtain a numerical integration of the diffusion equation. Not only is the simplicity of the analytical method lost in this alternative, but the values of

most of the parameters are not known with enough accuracy to justify the considerable additional complication. A more intuitively appealing variation is to select values of the principal parameters suitable to the conditions at each source, so as to more accurately determine concentrations near the source where they will be highest and hence most significant. (This we have tried in our sulphate calculation described below). Finally, it can be shown that variations of mixing height can be well approximated by selecting as the parametric value that at the receptor point.

Depletion effects are simulated by assuming a mass loss rate per unit volume of the form C/τ, where C is the mass concentration of the pollutant per unit volume and τ^{-1} is the first order rate constant for the loss of pollutant due to all causes. Three modes of depletion are taken into account: dry deposition, wet deposition caused by precipitation scavenging, and the loss of pollutant by chemical conversion of the primary species to a secondary form, (e.g., the conversion of SO_2 to sulfates). Thus the rate constant which takes into account all forms of depletion of a primary pollutant can be written as

$$\tau^{-1} = (\tau_d^{-1} + \tau_w^{-1} + \tau_c^{-1}) \tag{3}$$

where τ_d^{-1}, τ_w^{-1} and τ_c^{-1} are the dry deposition, wet deposition and chemical conversion rate constants respectively. Of course, only the chemical conversion term is truly a volume loss term, whereas the other depletion terms simulate the loss of pollutant to the ground, and must be suitably averaged through the mixing layer. Thus τ_c may be interpreted as the average time interval during which a molecule of primary pollutant drifts in the air before chemically converting to a secondary species.

Bearing these assumptions in mind, the horizontal distribution of concentration for a primary pollutant is determined by

$$u \frac{\partial C}{\partial x} + v \frac{\partial C}{\partial y} = D_h \left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2}\right) - \frac{C}{\tau} \tag{4}$$

where u and v are the components of the mean wind velocity in the x and y directions and D_h is the horizontal diffusivity.

Equation (4) can be solved for a steady point source having an emission rate Q and assuming the boundary condition of zero concentration at infinity. The horizontal distribution of concentration becomes:

$$C = \frac{Q}{2\pi h\, D_h} \exp\left(\frac{w}{2D_h} x'\right) K_o \left\{ r \left[\frac{1}{D_h \tau} + \left(\frac{w}{2D_h}\right)^2 \right]^{1/2} \right\} \tag{5}$$

where the x' axis is aligned in the direction of the mean wind velocity w, K_o is the modified Bessel function of the zeroth order, and r is the radial distance from the source to the receptor. It should be noted that (5) is a solution of a linear equation satisfying homogeneous boundary conditions. Thus, the principle of superpositioning the multiple sources applies, which we have utilized below.

Since (5) is a solution for a point source, we must disregard the solutions very near the source where predicted concentrations approach infinity. Because the function K_o is only weakly singular about r = 0, for (5) to be valid it is sufficient that r be large enough that the logarithm of the argument of K_o not be a large negative number. Also, the assumption of uniformly mixed pollutants within the mixing layer breaks down close to the source. In order to avoid the aforementioned difficulties we calculate concentrations at points no closer than about 10 kilometers from the source, a distance past which we assume that the pollutants have dispersed uniformly in the vertical direction.

The formation and dispersion of secondary pollutants can be treated by a diffusion model, similar to (4), which we now derive. Unlike primary pollutants which are emitted from point or area sources secondary products are formed continuously, within the atmosphere itself. The loss term in (4) due to chemical conversion of primary pollutant is proportionate to the source term in the secondary pollutant differential equation, namely $\beta C/\tau_c$, where C is the mass concentration of the primary pollutant, β is the mass ratio of secondary pollutant formed per unit mass of primary pollutant, and τ_c^{-1} is the rate constant for chemical conversion to a secondary species. Thus, the governing equation for the secondary pollutant becomes

$$w \frac{\partial C_s}{\partial x'} = D_h \left(\frac{\partial^2 C_s}{\partial x'^2} + \frac{\partial^2 C_s}{\partial y'^2} \right) - \frac{C_s}{\tau_s} + \frac{\beta C}{\tau_c} \qquad (6)$$

where C_s is the concentration of the secondary pollutant and $\tau_s^{-1} = (\tau_q^{-1} + \tau_r^{-1})$ is the net rate constant for loss of the secondary pollutants due to wet and dry deposition, whose respective rate constants are τ_q^{-1} and τ_r^{-1}.

Equation (6), although nonhomogeneous, may be solved in closed form for a distributed source of secondary pollutants formed from a point source of primary pollutant at r = 0. Thus the horizontal concentration profile becomes

$$C_s = \frac{\beta Q}{2\pi h D_h^2 \tau_c} \exp\left(\frac{w}{2 D_h} x'\right) \frac{K_o(\gamma r) - K_o(\alpha r)}{\alpha^2 - \gamma^2} \qquad (7)$$

where

$$\alpha^2 \equiv \left(\frac{1}{\tau_s D_h} + \frac{w^2}{4 D_h^2} \right) \tag{8}$$

$$\gamma^2 \equiv \left(\frac{1}{\tau D_h} + \frac{w^2}{4 D_h^2} \right) \tag{9}$$

Again, solutions due to multiple sources of primary pollutant can be superposed because the boundary condition at infinity specifies an ambient concentration of zero.

Considering the forms of (5) and (7), the spatial distribution of primary pollutant concentration C is characterized by two length scales, $\ell_1 = 2 D_h/w$ and $\ell_2 = (D_h \tau)^{1/2}$, while that of secondary pollutant C_s involves as well a third length scale $\ell_3 = (D_h \tau_s)^{1/2}$. The estimates of D_h given above lead to the conclusion that ℓ_1 is of the order of 10^6 m. (It will be seen below that ℓ_2 and ℓ_3 have the same magnitude for sulfur oxide dispersion). Thus the characteristic scale of the concentration pattern from a point source will be of the order of 10^6 m.

SECTION 3

COMPARISONS WITH TRAJECTORY-PLUME MODEL CALCULATIONS

As a first step in assessing the accuracy of the diffusion model for primary pollutants we compare our results with those of the trajectory-plume model developed by Heffter et al., (1975). For the purpose of this comparison, Heffter provided us with a sample computer calculation of the two-dimensional model which assumes a spatially uniform mixing height with pollutants well mixed in the vertical direction. For a steady point source in Illinois, mean concentration levels were calculated for trajectories averaged over the month of March 1975. Trajectories were initiated four times daily at six hour intervals and tracked for a maximum period of ten days. Contour maps depicted ambient concentration levels for two cases, the first without depletion and the second with depletion due to dry and wet deposition. Also calculated were deposition contours and individual trajectories.

The input parameters and data used in the comparative diffusion calculation were the same as these used in the trajectory calculation. The deposition velocity parameter used in the trajectory calculations was converted to its equivalent dry deposition rate constant by the relation

$$\tau_d = h/v_d \tag{10}$$

where v_d is the dry deposition velocity. Likewise an equivalent wet deposition rate constant was derived from the relation

$$\tau_w = (C/\bar{C})h/EP \tag{11}$$

where C is the ambient concentration at ground level, which in this case is assumed uniform within the mixing layer, $\bar{C}$ is the mean concentration of the pollution in the column of air from the ground to the top of the rainbearing layer, P is the precipitation rate and E is an empirically derived scavenging ratio. Trajectory data at the source was used to calculate the values of the mean wind speed and direction. In these calculations various values of D_h were assumed and the diffusion model concentration contours were compared with these of the trajectory calculation. It was found that $D_h = 6.4 \times 10^5$ m^2/s gave the best overall agreement for all comparisons made. This value of the diffusion coefficient is near that given by Murgatroyd (1969) and Durst et al. (1959).

The comparative calculations for two-dimensional diffusion without and with depletion effects are shown in Figs. 1 and 2, respectively. For these calculations the mixing layer height was 1500 m, the mean wind velocity was 3.2 m/s at the point of emission with a wind direction from 265° true. For the calculation displayed in Fig. 2, depletion was assumed to have occurred by two mechanisms, dry deposition with a deposition velocity v_d = 0.01 m/sec and depletion due to precipitation scavenging with a rate constant $\tau_w = 3 \times 10^5$ sec. The net depletion time τ was consequently determined to be 10^5 sec. An obvious consequence of including depletion effects is that the contour profiles tend to decrease in area and also close at lesser distances downwind. Compared with Fig. 1, there is better agreement between the two models when depletion effects are included, as in Fig. 2.

Judging from the graphical comparison of Figs. 1 and 2, it is evident that there are some differences between the results of each model. In the case of no depletion it is specially evident that the trajectory model contours tend to close at lesser distances downwind from the source than the diffusion model contours. In the trajectory model, trajectories are computed for a maximum of ten days travel time so as not to exceed computer capacity limitations. Thus, regions outside the range of these trajectories experience zero values for the ambient concentration. The diffusion model, however, desribes a steady state situation where the ambient concentration approaches zero at infinity, a condition that would probably be approximated by the trajectory model if the individual trajectories were followed for a infinite time interval and to an infinite distance.

It is also evident that the trajectory model exhibits puckering of the contour lines while the diffusion model exhibits smooth contours that are symmetric about the axis of the direction of the mean wind velocity. The puckering probably is a consequence of averaging concentrations along trajectories for the relatively short period of one month, for which 120 trajectories were not fully random in character and some wind directions were more likely than others, causing some of the trajectory paths to be grouped and contour profiles to become puckered. It is believed that averaging concentrations for a longer period of time (e.g. annually) would result in "smoother" contour profiles because the trajectories would not be lined up in any preferred direction.

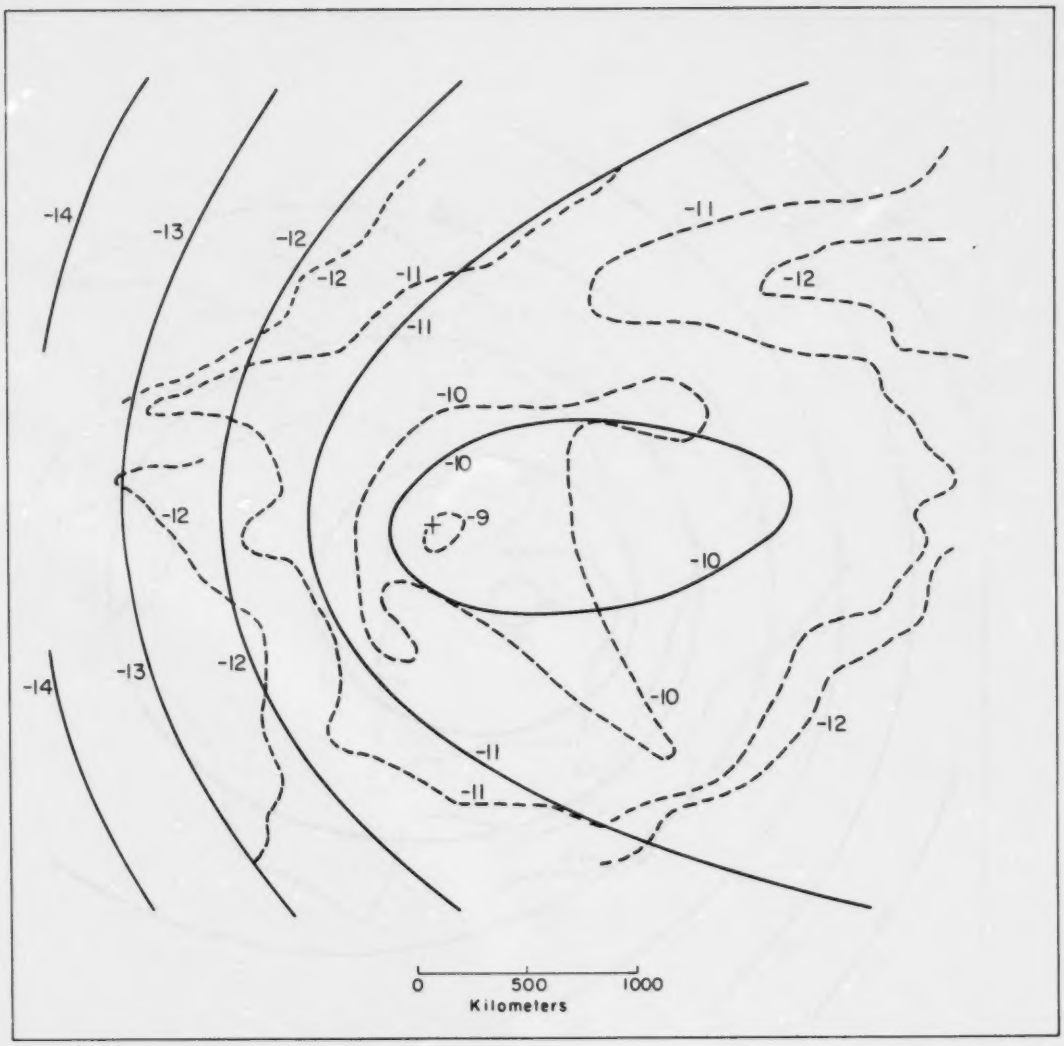

Figure 1. Comparison of model calculations of isopleths of primary pollutant concentration for a point source (+) without depletion effects. Dashed lines are determined from a plume-trajectory model and solid lines from the analytical diffusion model. Source strength is 1 kg/s. Isopleth value is the logarithm of the concentration (kg/m^3).

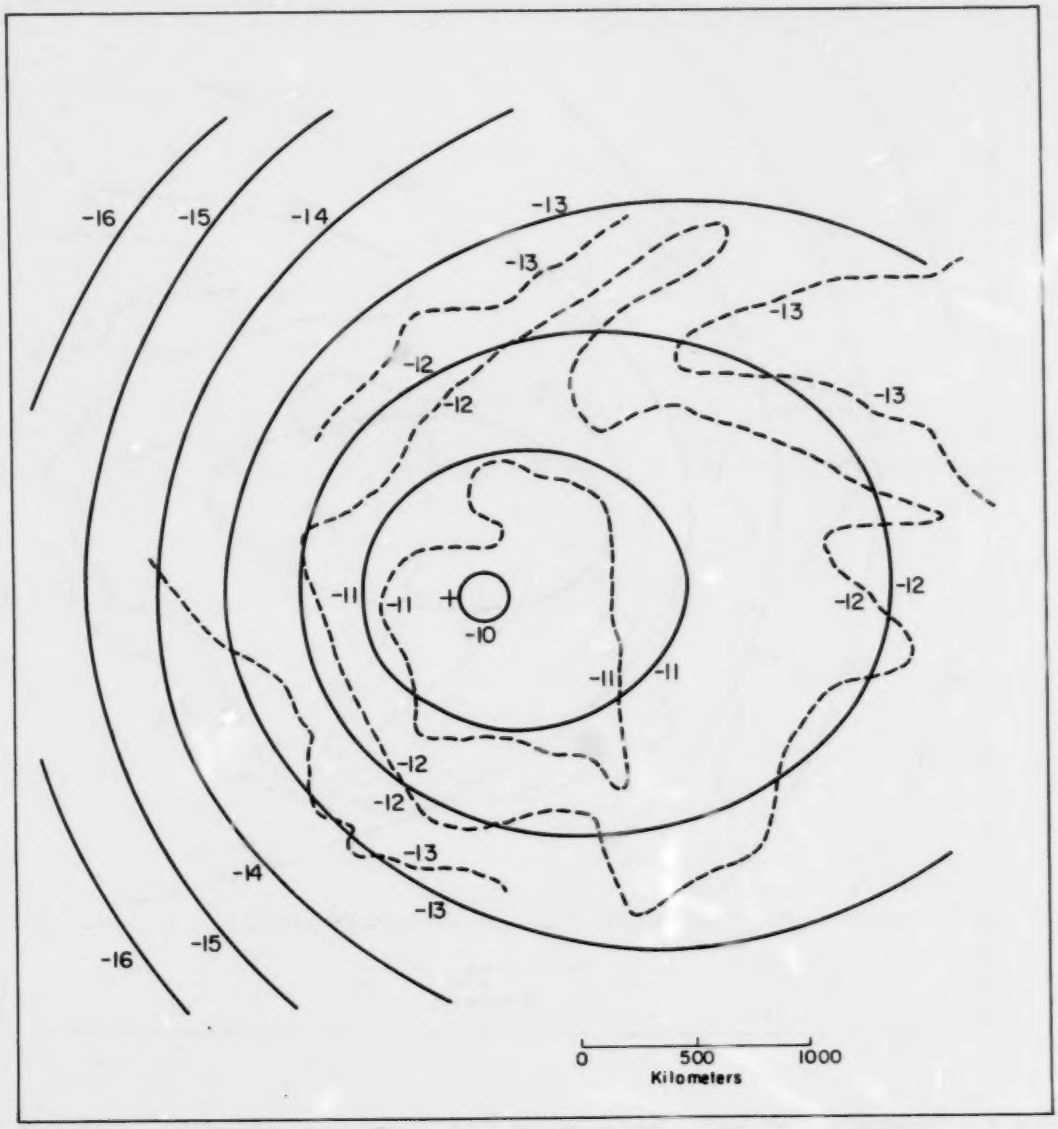

Figure 2. Same comparison as in Figure 1, but including depletion by wet and dry deposition.

SECTION 4

COMPARISON WITH U.S. ATMOSPHERIC SULFATE MEASUREMENTS

Because most current interest has centered on the long distance transport of sulfur compounds, we have compared the results of our model calculations with measurements of atmospheric sulfate concentrations within the continental U.S. In making this comparison, we have assumed that the major contributors to ambient sulfate levels are the elevated sources of sulfur dioxide, following the conventional argument that sulfur dioxide emitted from low or gound level sources is significantly depleted close to the source by deposition to the ground before it can be converted to sulfate in the atmosphere. As a convenient elevated source inventory we use the emissions from fossil fuel electric power plants, which comprise the major proportion of such emissions. However, in using our model for the purpose of this comparison, we assume that there exists a geographically uniform background level of sulfates whose level is to be determined empirically, added to which is the contribution from the power plant emissions. Low level anthropogenic and biogenic SO_2 emissions can thus be considered as the principal source of this sulfate background.

As there are about 700 power plants to be included in this inventory, we adopted the following simplification in order to reduce the length of the computation. The power plant emissions, as estimated by the Federal Power Commission (1973) for the year 1970, were aggregated within each air quality control region, as defined by the Environmental Protection Agency (1972). These aggregated emissions were then considered to issue from a single source located in the center of each air quality control region. Our aggregated inventory thus consisted of 106 point sources comprising 96% of total U.S. power plant emissions, the remainder being judged too small to justify their inclusion.

An advantage of our model is the relative ease with which multiple sources may be superposed. Trajectory-plume model calculations for 700 power plants, or even 100 air quality control region sources, would be very expensive, especially if averaged over a season or year. Some aggregation of the sources, as we have done, is not essential to the use of the model, but is a practical convenience having negligible effect on the precision of the results.

Using trial values for the parameters in equation (7), a calculation was made of the annual ambient sulfate concentrations for comparison with the measurements described by Hidy et al. (1976), which comprise a network of 222 stations located as shown in Fig. 3. A disappointingly low correlation coefficient of 0.46 was obtained between the observations and the model predictions. Our model could not account for the significant variations in concentrations within short distances which were observed.

Others have encountered similarly poor correlations. Wendell et al. (1977) compared trajectory-puff model calculations with these same measurements averaged for the month of April 1974, finding a correlation coefficient of 0.40. Similarly, the spatial variability of observed data could not be duplicated by their model. They suggested that uncertainties in the observed data might be responsible for the poor correlation. For example, they noted that most air quality monitoring stations were located in urban areas where the depletion rates might be higher than in rural areas and that SO_2 interference on high volume samplers may introduce errors of unknown amount in the sulfate measurement. In addition to these suggested effects, we believe that the significant spatial variability in the observed data may be also a result of locally induced phenomena such as the unique terrain features upon which the various sensing instruments are located and sulfur emissions from small local sources which have not been accounted for in the model calculations.

In discussing the use of a trajectory-puff model to describe sulphur transport and deposition in western Europe, Eliassen (1978) noted that the correlation coefficient for model and measured average daily sulfate levels varied between 0.4 and 0.8, and showed that the annual average sulfate contours predicted by the model gave less spatial variability than the measured values.

Those effects which give rise to local variability in the measurements may be diminished by smoothing out the measured data. Accordingly, we fitted, by least squares, a polynomial surface to the data points, using latitude and longitude as independent variables. A best correlation coefficient of 0.75 was obtained for a fifth order polynomial when comparing the measured data with the polynomial representation. In all of the subsequent comparisons with our model, the smoothed representation of the measured data was used.

In our calculations the mixing height h used in (7) was evaluated at each receptor point as the harmonic mean of the values given by Holzworth (1972) for mean annual morning and afternoon mixing heights. At most, this variation did not exceed 25% of a national average of about 650 m.

In the first comparisons between the model calculation and observation with respect to the spatial distributions of concentrations across the U.S., it was noticed that the west coast region might be better described by a set of parameters which differ from those in the east. Therefore, we divided the United States into two separate regions at the 100th meridian and attempted to correlate the model with the data separately for each region. The east contained 173 data points while the west had 49.

As previously mentioned, the solution (7) for the spatial distribution of secondary pollutant concentration depends parametrically upon three dimensional length scales. In addition, the wind direction and the scaling factor $D_h^2 \tau_c$ are also parameters, the mixing height being determined as described above. Concentrating on the eastern U.S., we varied these five parameters so as to maximize the correlation coefficient for the model calculation as compared with the smoothed representation of the data, obtaining a value of 0.87 when using the optimum choice of dimensional parameters listed in Table 1. A background level of 5.95 $\mu g/m^3$ was obtained from this linear regression.

For the western half of the U.S. a smaller correlation coefficient was obtained (0.69) and the optimum dimensional length scales were smaller (Table 1) as might be expected from the higher gradients of concentration observed on the southwest coast where most measuring stations are located. In an alternate trial, using a different west coast source inventory compiled by the U.S. Environmental Protection Agency (1974) for the year 1972, a still lower correlation coefficient of 0.53 was found. This particular inventory included estimates of emissions from all sources, not just those from power plants.

Figure 3 depicts the diffusion model concentration contours (solid lines) resulting from use of the optimal values of the parameters for the eastern and western United States as given in Table 1. Also shown are the contours of the smoothed sulfate data (dashed lines) and the location of sulfate monitoring stations. It can be seen that good agreement, both quantitative and qualitative, is achieved in the eastern portion of the United States. In the western portion, however, agreement is less favorable. In southern California especially peak values of the ambient concentration are underpredicted while the contrary is true for the northwest mountain region. These less favorable results in the west may be due in part to our arbitrary division of the United States at the hundredth meridian into eastern and western regions and in part to real effects of regional topography. In addition, since there are fewer measuring stations in the western region, the correlation there is less significant than in the east.

The sulfate background level of about 6 $\mu g/m^3$ (Table 1) determined from the linear regression is a little higher than values reported for rural regions. Ottar (1978) notes that annual sulfate concentrations in western Europe are about 1 $\mu g/m^3$ in the more remote (western) regions. Meszaros (1978) summarizes values of 3 to 5 $\mu g/m^3$ for rural western and eastern U.S. sites. Recognizing the spatial variability of the measurements, the agreement is satisfactory.

The four optimum dispesion parameters listed in Table 1 incorporate five physical parameters: w, D_h, τ, τ_c and τ_s. While these latter values are thus not determinable from the four dispersion parameters, we suggest a set of values for them, shown in Table 2, which is consistent with Table 1 and various estimates given in the literature. Also given in Table 2 are specific estimates for or bounds upon these values as given by other investigators. Of these, the values of Johnson et al. (1978) and Eliassen (1978) were suggested by examination of the comparison of trajectory model calculations with observations. There is general agreement among the suggested values for the significant time scales. The advection parameters, w and D_h, are used in our model to replace trajectory calculations.

Additional model calculations were carried out for the eastern U.S. illustrating several interesting points. A calculation using optimal parameters as determined previously but assuming a uniform mixing height resulted in a correlation coefficient of 0.87, equal to that for the case incorporating variable mixing height. Another calculation using the same optimal parameters as above and a 1972 inventory of all sulfur sources (U.S. Environmental Protection Agency 1974) gave an equal correlation coefficient of 0.87. We thus conclude that the model results are not sensitive to the spatial distributions of mixing height (and probably other dispersion parameters as well) and source inventory.

TABLE 1 OPTIMUM SULFATE DISPERSION PARAMETERS

	Eastern U.S.	Western U.S.
τD_h (m^2)	1.3×10^{11}	1.0×10^{9}
$\tau_s D_h$ (m^2)	2.5×10^{11}	1.3×10^{9}
D_h^2/w^2 (m^2)	4.0×10^{12}	6.4×10^{9}
Wind Direction	270° true	270° true
$D_h^2 \tau_c$ (m^4/s)	9.78×10^{17}	1.02×10^{13}
Correlation Coefficient	.87	.69
Background ($\mu g/m^3$)	5.95	5.65

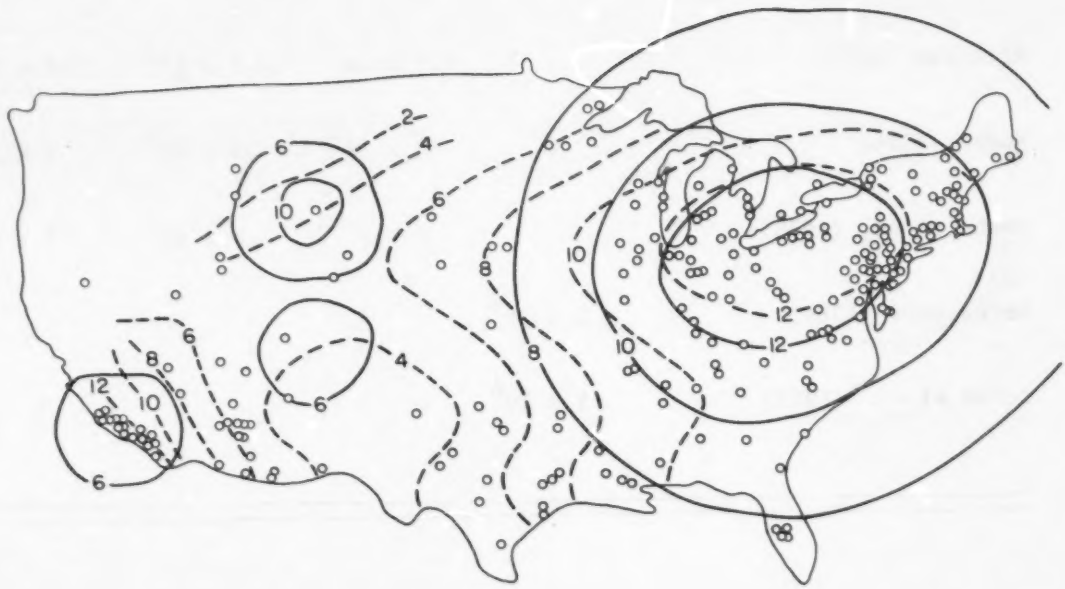

Figure 3. A comparison of calculated (solid line) and measured (dashed line) annual average sulfate concentrations ($\mu g/m^3$) in the U.S. Small circles give location of monitoring stations. Measured isopleths are determined from a smoothed correlation of the raw data.

TABLE 2 SULFATE DISPERSION CONSTANTS

	w (m/s)	D_h (m^2/s)	τ (s)	τ_s (s)	τ_c (s)
This study, eastern U.S.	2.0	2.0×10^6	8×10^4	1.6×10^5	2.4×10^5
Johnson et al. (1978)			$<5 \times 10^5$	$<5 \times 10^5$	3.6×10^5
Garland (1978)			$<10^5$	$>10^4$	4×10^5
Eliassen (1978)			2.5×10^4	2.5×10^5	2.8×10^5
Rodhe (1978)			9×10^4	3×10^5	3×10^5
Prahm et al. (1976)			5×10^4	2.5×10^5	3×10^5
Murgatroyd (1969)		2×10^6			
Durst et al. (1959)		5×10^6			

As can be seen in Fig. 3, the contours of annual average sulfate concentration show only gradual changes over large distances. Eliassen (1978) also notes this consequence of his transport model. In both instances, since the secondary pollutant sulfate is produced over a broad area, even when the primary pollutant sulfur dioxide issues from a point source, it is to be expected that no small regions of high local concentration will exist.

A more precise statement can be made by examining the nature of the solution (7) for the secondary pollutant concentration for a single point source of primary pollutant. For radial distances r which are small compared with the length scales $2D_h/w$, α^{-1} and γ^{-1} the maximum value $(C_s)_m$ of C_s is found to be:

$$(C_s)_m = \frac{\beta Q \ln(\alpha/\gamma)}{2\pi h D_h^2 \tau_c (\alpha^2 - \gamma^2)}$$

$$= [3 \times 10^{-11} \text{ s/m}^3] Q \qquad (12)$$

where the numerical value follows from the use of the parameter values given in Table 2 for sulfate dispersion. What is also evident from an examination of the form of (7) is that C_s does not decrease by a factor of two below this maximum until r exceeds about 500 km. This is in marked contrast to the substantial reduction in primary pollutant concentration within a comparable distance from the source (see Fig. 2, for example). It is also interesting to note from (12) that a single source (or a group of closely spaced sources) of 33 kg/s of sulfur dioxide produces an average increment of 1 $\mu g/m^3$ of sulfate within this large region.

Finally, we determined the annual average SO_2 concentration from equation (5) using the optimum dispersion parameters of Table 1 and the power plant source inventory. The resulting diffusion model concentration contours are illustrated in Fig. 4 (smooth lines) along with approximate contours derived from measurements of annual average SO_2 levels in urban areas for 1970-71 (dashed lines) as depicted by the National Academy of Sciences (1975). The diffusion model contours are similar in shape to the measured values, but tend to overpredict ambient concentrations in the northeastern U.S. A more careful analysis and comparison of this case seems warranted. It is interesting to note that Eliassen (1978) also found that his model calculations give a higher ratio of sulfur dioxide to sulfate than was observed, as is evident from comparing our Figs. 3 and 4.

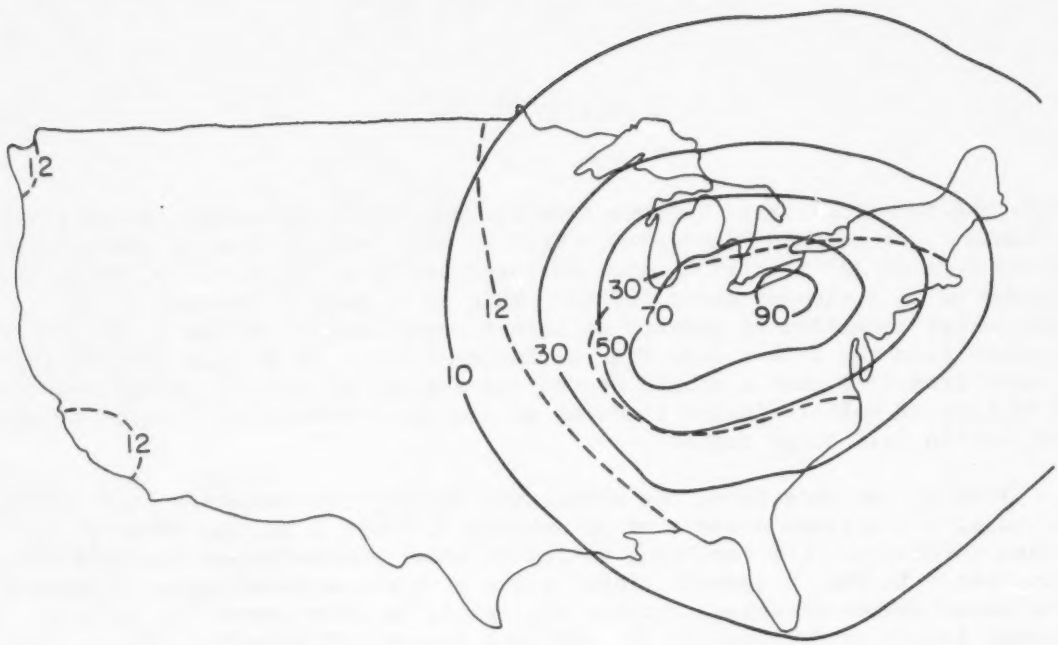

Figure 4. A comparison of annual average SO_2 concentrations ($\mu g/m^3$) determined from the analytical diffusion model (solid lines) and measurements (dashed lines) for the same model parameters as were used in obtained the sulfate concentrations of Fig. 3.

SECTION 5

CONCLUSIONS

Our diffusion theory for the dispersion of primary and secondary pollutants shows promise as an alternate and simple analytical method for evaluating the time-averaged effects of long distance pollutant transport on regional air quality. By assuming that large scale wind fluctuations act in a random manner over a sufficiently long period of time and that depletion and transformation processes are of first order, we find analytical solutions for the concentrations of both primary and secondary pollutants. For multiple point sources, these solutions can be superposed. Thus the computational effort involved in multiple source calculations is only a fraction of that necessary for the analogous trajectory calculation.

Limited calculations using the diffusion model for a single point source have been shown to be in good overall agreement with a more detailed trajectory calculation when the horizontal diffusivity is appropriately selected and identical values of other parameters are used. This resulting diffusivity was found to be comparable with values derived from other trajectory studies.

Comparisons of our model calculation of the annual average concentrations of sulfate formed from U.S. fossil fuel power plant emissions with ambient measurements yield a good correlation when model parameters are optimally chosen and the measured data are smoothed to reduce variations on a small geographic scale. These comparisons appear to be as good as those obtained from trajectory models describing sulfur transport in western Europe. The optimum values of the diffusion model parameters we have determined are consonent with the rate constants for sulfur oxide atmospheric behavior and diffusivities as reported in the literature.

REFERENCES

Cleveland, W.S., Kleiner, B., McRae, J.E. and Warner, J.L. (1976) Photochemical air pollution: transport from the New York City area into Connecticut and Massachusetts. Science, 101, 197.

Draxler, R.R., (1976) A diffusion deposition scheme for use within the ARL trajectory model. NOAA TM ERL ARL 63, U.S. Department of Commerce, National Oceanic and Atmospheric Administration, Washington, D.C.

Durst, C.S., Crossley, A.F. and Davis, N.E., (1959) Horizontal diffusion in the atmosphere as determined by geostrophic trajectories. J. Fluid Mech., 6, 401.

Eliassen, A., (1978) The OECD study of long range transport of air pollutants: long range transport modelling. Atmospheric Environment, 12, 479.

Eliassen, A., and Saltbones, J., (1975) Decay and transformation rates of SO_2 as estimated from emission data, trajectories and measured air concentrations. Atmospheric Environment, 9, 425.

Federal Power Commission, (1973) Steam electric plant air and water quality control data for the year ended December 31, 1970, Washington, D.C.

Garland, J.A., (1978) Dry and wet removal of sulfur from the atmosphere. Atmospheric Environment, 12, 349.

Gillani, N.V. and Husar, R.B., (1976) Mesoscale model for pollutant transport, transformation and ground removal. Presented at the third symposium on atmospheric turbulence, diffusion and air quality, October 19-22, 1976, Raleigh, North Carolina.

Glotfelty, D.E. (1978) The atmosphere as a sink for applied pesticides. J. Air Pollution Control Assoc., 23, 917.

Heffter, J.L., Taylor, A.D. and Ferber, G.J., (1975) A regional continental scale transport diffusion and deposition model. NOAA TM ERL ARL 50, U.S. Department of Commerce, National Oceanic and Atmospheric Administration, Washington, D.C.

Hidy, G.M., Mueller, P.K. and Tong, E.Y. (1978) Spatial and temporal distributions of airborne sulfate in parts of the United States. Atmospheric Environment, 12, 735.

Hidy, G.M., Tong, E.Y. and Mueller, P.K., (1976) Design of the sulfate regional experiment (SURE). EPRI EC-125, Electric Power Research Institute, Palo Alto, California.

Holzworth, C.G., (1972) Mixing heights, wind speeds and potential for urban air pollution throughout the contiguous United States. AP-101, U.S. Environmental Protection Agency, Research Triangle, N.C.

Johnson, W.B., Wolf, D.E. and Mancuso, R.L. (1978) Long term regional patterns and transfrontier exchanges of airborne sulfur pollution in Europe. Atmospheric Environment, 12, 511.

Meszaros, E., (1978) Concentration of sulfur compounds in remote continental and oceanic areas. Atmospheric Environment, 12, 699.

Murgatroyd, R.J. (1969) Estimations from geostrophic trajectories of horizontal diffusivity in the mid-latitude troposphere and lower stratosphere. Q. Jl. R. Met. Soc., 95, 40.

National Academy of Sciences, (1975) Air quality and stationary source emission control. Serial No. 94-4, Committee on Public Works, U.S. Senate, U.S. Government Printing Office, Washington, D.C.

National Research Council, (1978) Nitrates: an environmental assessment. Nation National Academy of Sciences, Washington, D.C.

Ottar B., (1978) An assessment of the OECD study on long range transport of air pollutants (LRTAP). Atmospheric Environment, 12, 455.

Pasquill, F., (1974) Limitations and prospects in the estimation of dispersion of pollution on a regional scale. Advances in Geophysics, 18B, 1.

Prahm, L.P., Torp, U. and Stern, R.M., (1976) Deposition and transformation rates of sulfur oxides during atmospheric transport over the Atlantic. Tellus, 28, 4.

Rodhe, H., (1978) Budgets and turn-over times of atmospheric sulfur compounds. Atmospheric Environment, 12. 671.

Sheih, C.M., (1977) Application of a statistical trajectory model to the simulation of sulfur pollution over northeastern United States. Atmospheric Environment, 11, 1973.

Shirvaikar, V.V., (1972) Persistence of wind direction. Atmospheric Environment, 6, 889.

Start, G.E. and Wendell, L.L., (1974) Regional effluent dispersion calculations considering spatial and temporal meteorological variations, NOAA TM ERL ARL 44, U.S. Department of Commerce, National Oceanic and Atmospheric Administration, Washington, D.C.

Symposium on sulfur in the atmosphere (1978). Atmospheric Environment, 1-3, 1.

Taylor, G.I., (1921) Diffusion by continuous movements. Proc. London Math. Soc., 20, 196.

U.S. Environmental Protection Agency, (1972) Federal air quality control regions. AP-102. Rockville, Md.

U.S. Environmental Protection Agency, (1974) 1972 National emissions report. EPA-450/2-74-012, Research Triangle Park, N.C..

Wendell, L.L., Powell, D.C. and McNaughton, D.J., (1977) A multi-source comparison of the effects of real time versus time averaged precipitation data on SO_2 and sulfate particulate removal in a regional assessment model. Preprints Joint Conference on Applications of Air Pollution Meteorology, Salt Lake City, Utah.

Wolff, G.T., Lioy, P.J., Wight, G.D., Meyers, R.E. and Cederwall, R.T., (1977) An investigation of long-range transport of ozone across the midwestern and eastern United States. Atmospheric Environment, 11, 797.